

**REMARKS**

Applicants have amended their claims in order to further clarify the definition of various aspects of the present invention. Specifically, Applicants have incorporated the subject matter of claim 17 into claim 1 (that is, have amended claim 1 to recite that the temperature at which the reaction of the aromatic compound represented by the general formula (1) with the olefin is performed, is not higher than 20°C but not lower than -20°C). In light of amendments to claim 1, claim 17 has been cancelled without prejudice or disclaimer; and, moreover, claims 3 and 9 have also been cancelled without prejudice or disclaimer. In addition, dependencies of claims 13 and 18 have been amended, respectively to be dependent on claims 11 and 1.

The rejection of claim 13 under the second paragraph of 35 USC 112, as being indefinite, set forth in Item 4 of the Office Action mailed March 13, 2008, is moot, in light of amendment of dependency of claim 13 to be dependent on claim 11, rather than on cancelled claim 12.

Applicants respectfully submit that the claims presented for consideration by the Examiner patentably distinguish over the teachings of the references applied by the Examiner in rejecting claims in the Office Action mailed March 13, 2008, that is, the teachings of the U.S. patents to Olah, No. 3,766,286, and to Frey, No. 2,372,320, under the provisions of 35 USC 102 and 35 USC 103.

It is respectfully submitted that these references as applied by the Examiner would have neither taught nor would have suggested such a process for producing an alkyl aromatic compound as represented by the general formula (2) in the present claims, including wherein an aromatic compound represented by the general formula

(1) as in the present claims ( $R^1$  and  $R^2$  of the general formula (1) being in the meta positions in the general formula (1) and each being a methyl group) is alkylated with an olefin having 2-4 carbon atoms in the presence of a Broensted acid and without presence of a Lewis acid, with the resulting mixture being subsequently added with a Lewis acid and being subjected to isomerization in the copresence of the Broensted acid and the Lewis acid, the alkylation step being performed at a temperature in the range of  $-20^{\circ}$  to  $20^{\circ}\text{C}$  (that is, at a relatively low temperature), and with the Broensted acid being HF and the Lewis acid being  $\text{BF}_3$ . Note claim 1.

Furthermore, it is respectfully submitted that these applied references would have neither taught nor would have suggested such a process for producing an alkyl aromatic compound represented by the general formula (2), as in the present claims, having features as discussed previously in connection with claim 1, and, in particular, wherein X is a hydrogen atom in the general formulas (1) and (2), as in claims 8 and 14, with  $R^0$  being an isopropyl group in the general formula (2), as also in claims 8 and 14; or wherein X is a methyl group (see claim 22).

As can be appreciated, where  $R^1$  and  $R^2$  are a methyl group and X is a hydrogen atom, the starting material is m-xylene; and where  $R^1$  and  $R^2$  are each a methyl group and X represents a methyl group also, the starting material is a trimethylbenzene (e.g., 1, 2, 4-trimethylbenzene) in the alkylation reaction.

As will be discussed in more detail infra, Frey discloses that the alkylatable hydrocarbon is illustratively benzene, but it is also within the scope thereof to alkylate toluene or isoparaffins such as isobutane or isopentane; and it is respectfully submitted that this reference, either alone or in combination with the teachings of Olah, would have neither disclosed nor would have suggested such process as in

the present claims, wherein a starting material for the alkylation is an aromatic compound represented by the general formula (1) wherein R<sup>1</sup> and R<sup>2</sup> are each a methyl group and are in meta positions, or wherein the alkylation is carried out in the absence of a Lewis acid.

Furthermore, and as will also be discussed further infra, noting that Frey discloses relatively high temperatures for the alkylation process described therein, it is respectfully submitted that the combined teachings of the applied references would have taught away from such process as in the present claims, wherein the alkylation step is conducted at a temperature not higher than 20°C but not lower than -20°C.

In addition, it is respectfully submitted that the teachings of these applied references would have neither disclosed nor would have suggested such process as in the present claims, having features as discussed previously in connection with claim 1, and, moreover, the molar ratios of various components in the reaction isomerization as in claims 2, 4, 5, 10, 15 and 16; and/or temperature of the isomerization as in claims 5, 11 and 18-20, especially together with the temperature of the alkylation as in claim 1; and/or wherein the olefin is selected from the group as set forth in claims 7 and 13; and/or wherein the alkylation and the isomerization are carried out in a same single reaction chamber (see claim 21).

The present invention relates to a process for producing an alkyl aromatic compound of the general formula (2) in claim 1, from a specified aromatic (benzene-ring containing) compound (formula (1) in claim 1) having two methyl substituents in meta positions, by, inter alia, alkylating the aromatic (benzene-ring containing) compound of general formula (1) in claim 1, having two methyl substituents in meta

positions, with a lower olefin such as ethylene or propylene. The produced alkyl aromatic compound is useful as a raw material for various industries, such as in pharmaceuticals, agricultural chemicals, liquid crystals, functional pigments, solvents and monomers for engineering plastics.

Various techniques have been used for the alkylation, as described on pages 1-3 of Applicants' specification. These alkylation reactions use, as a catalyst, a Lewis acid represented by aluminum chloride; and also well known is an alkylation reaction using a catalyst HF which is a Broensted acid. However, these previously proposed methods have various problems, including a problem in connection with separation and purification of HF used as the catalyst, and wherein the yield of the desired product is undesirably low, or conversion of the raw material is not satisfactory.

In alkylating a substituted benzene compound having two methyl substituents on the benzene ring, the position (ortho, meta or para position) on the aromatic ring of the raw material, to which an alkyl group is introduced, is determined by the effect of the functional groups on the reactant substituted benzene compound. When the desired compound is not in accord with the substitution orientation inherent to the functional group, however, a positional isomer of the desired compound is obtained as a product, and it is necessary to carry out a disproportionation reaction, called an isomerization reaction or a transalkylation reaction, in order to introduce the alkyl group to the desired position.

Against this background, Applicants provide a process for producing an alkyl benzene compound having substituents at the 3- and 5-positions (meta positions), which is high in yield and in selectivity to the desired compound, and which permits

easy and efficient recovery and recycling of the catalyst. As described on page 5 of Applicants' specification, the present inventors have found that the desired alkyl benzene compound having substituents at the 3- and 5-positions can be obtained in a stable manner with a high yield and a high selectivity, under mild and simple reaction conditions, by (1) alkylating a benzene compound having two substituents at the meta positions with an olefin having 2-4 carbon atoms in the presence of a Broensted catalyst such as HF, and without the presence of a Lewis acid, followed by (2) addition of a Lewis acid such as  $\text{BF}_3$  and permitting intramolecular isomerization to proceed.

Applicants have found that a Lewis acid hinders the alkylation reaction, and, therefore, should not be used in the alkylation stage. In view thereof, Applicants provide a process achieving both effective alkylation and isomerization, by performing the alkylation in the presence of the Broensted acid and without the presence of the Lewis acid, with the Lewis acid subsequently being added and wherein the isomerization takes place in the copresence of the Broensted acid and the Lewis acid. Note the last full paragraph on page 7, and the paragraph bridging pages 7 and 8, of Applicants' specification.

As to advantages achieved according to the present invention, attention is respectfully directed to the Examples and Comparative Examples on pages 10-15 of Applicants' specification. This evidence in Applicants' specification must be considered in determining obviousness. See In re DeBlauwe, 222 USPQ 191 (CAFC 1984).

As can be seen in Comparative Examples 2-7 and as discussed in the last full paragraph on page 13 of Applicants' specification, it is seen that, when  $\text{BF}_3$  is added

in the alkylation reaction stage, good results including a high yield and high selectivity are not obtained, even when amounts of HF and propylene are changed. Note also the results discussed in connection with Comparative Example 8 on page 14 of Applicants' specification, wherein it is described that the product obtained had a yield of desired 3,5-dimethylcumene of 7%, and the selectivity was 13% and the yield of 2,4-dimethylcumene was 27%. In the Examples according to the present invention, much higher yield and selectivity were achieved.

It is respectfully submitted that the unexpectedly better results achieved according to the present invention, as seen in comparing the Examples and Comparative Examples in Applicants' specification, clearly support unobviousness of the presently claimed invention, wherein the alkylation is performed in the presence of a Broensted acid and without the presence of a Lewis acid, with a Lewis acid subsequently being added and isomerization taking place in the copresence of the Broensted acid and the Lewis acid.

In the Office Action mailed March 13, 2008, in Item 11 thereof, the Examiner contends that the evidence of record does not provide a comparison with the closest prior art. It appears that the Examiner is requiring a comparison with the combination of teachings of Frey and Olah. Note the second paragraph of Item 11 of the Office Action mailed March 13, 2008. It is respectfully submitted that any requirement for comparison with the combination of teachings of the applied references is improper. See Manual of Patent Examining Procedure (MPEP) 716.02(e), sub-heading III, as well as sub-heading I.

Olah discloses a process for isomerizing paraffins and/or alkyl substituted aromatic hydrocarbons, the isomerizable hydrocarbons being selected from the

group consisting of paraffins, alkyl substituted aromatic hydrocarbons, and mixtures thereof, the hydrocarbons being isomerized at low temperatures in the presence of a catalyst comprising (a) one or more Lewis acids of a specified formula, and (b) a strong Bronsted acid, preferably comprising a strong halogen-substituted acid such as fluorosulfuric acid, trifluoromethanesulfonic acid, trifluoroacetic acid or mixtures thereof. Note the paragraph bridging columns 1 and 2 of Olah. See also column 3, lines 50-52.

It is respectfully submitted that Olah would have neither disclosed nor would have suggested alkylation of an aromatic compound represented by the general formula (1) as in the present claims, much less wherein such alkylation is performed at the relatively low temperature in a range not higher than 20°C but not lower than -20°C, with such alkylation being performed in the presence of a Bronsted acid and without the presence of a Lewis acid, together with the isomerization, and advantages achieved thereby particularly with respect to higher yield and selectivity achieved.

It is respectfully submitted that the additional teachings of Frey would not have rectified the deficiencies of Olah, such that the presently claimed invention as a whole would have been obvious to one of ordinary skill in the art.

Frey discloses a process for the catalytic alkylation of hydrocarbons. This patent discloses admitting an alkylatable hydrocarbon to an alkylator, with concentrated or substantially anhydrous hydrofluoric acid being admitted. This patent discloses that for purposes of illustration benzene is chosen as the alkylatable hydrocarbon; but it is within the scope of the process described in the patent to alkylate also toluene or iso-paraffins such as isobutane or isopentane. All examples

of this patent use benzene. See page 2, left-hand column, lines 13-22. Note also page 3, right-hand column, lines 22-31.

It is respectfully submitted that Frey does not expressly disclose that the alkylation is carried out in the absence of a Lewis acid.

Initially, it is emphasized that Olah discloses an isomerization process, while Frey discloses an alkylation process. Specific isomerizable hydrocarbons are described in connection with Frey, and are not disclosed in Olah. The combined teachings of Frey and Olah do not disclose, nor would have suggested, a need for the isomerization process after the alkylation process of Frey. It is respectfully submitted that one of ordinary skill in the art concerned with in Olah, pertaining to an isomerization reaction, would not have looked to the teachings of Frey, disclosing specific compounds not specifically set forth in Olah, and disclosing an alkylation process; in particular, there would have been no reason from the combined teachings of these references, or in any other manner to one of ordinary skill in the art, to combine the teachings of these references. It is respectfully submitted that only through hindsight use of Applicants' disclosure, which is improper under 35 USC 103, would one of ordinary skill in the art have combined the teachings of Olah and Frey, as applied by the Examiner.

In any event, even assuming, arguendo, that the teachings of Olah and Frey were properly combinable, such combined teachings would have neither disclosed nor would have suggested the combination of alkylation and isomerization steps as in the present claims, of the aromatic compound presented by the general formula (1) as in claim 1, wherein R<sup>1</sup> and R<sup>2</sup> are each a methyl group, at meta positions as discussed previously, particularly the aromatic compound as in claims 8 and 14,



wherein, additionally, X is H, or the aromatic compound as in claim 22, wherein X is a methyl group, the alkylation being performed at a temperature as in claim 1, with the alkylation being performed in the presence of a Broensted acid and without the presence of a Lewis acid, while the isomerization is carried out in the copresence of these acids, and advantages thereof; and/or other features of the present invention as recited in the present claims, including, inter alia, both processing steps are carried out in a same single reaction chamber, and advantages thereof.

It is acknowledged that Frey discloses at page 2, left-hand column, lines 20-22 that "it is within the scope of this invention to alkylate also toluene or isoparaffins such as isobutane or isopentane". Such disclosure in Frey, describing specific materials to be alkylated "within the scope of this invention" [in Frey], would have taught away from the presently claimed process, including alkylation of the aromatic compound represented by the general formula (1) in claim 1, wherein R<sup>1</sup> and R<sup>2</sup> are each a methyl group, at meta positions, as in all of the present claims, much less wherein X is a hydrogen atom (see claims 8 and 14) or wherein X is a methyl group (see claim 22).

It is emphasized that Frey discloses that the invention therein is directed to benzene, toluene or isoparaffins such as isobutane or isopentane as the alkylatable hydrocarbon. Such disclosure in Frey, even in combination with the teachings of Olah, would have taught away from the present invention, wherein the aromatic compound represented by the general formula (1) has each of R<sup>1</sup> and R<sup>2</sup> being a methyl group, at meta positions.

Moreover, note that in Frey an illustrative temperature range for the alkylation, using benzene as the alkylatable hydrocarbon, is generally in the range of 70°-

120°F. Such relatively high temperature range for the alkylation in Frey has a minimum temperature that is greater than the maximum temperature of the temperature range in the present claims. It is respectfully submitted that the combined teachings of Olah and of Frey would have neither disclosed nor would have suggested, and in fact would have taught away from, the temperature range for the alkylation as in all of the present claims.

The contention by the Examiner in the eighth paragraph of Item 8 of the Office Action mailed March 13, 2008, that it would have been obvious to modify, inter alia, the temperature range for alkylation, is respectfully traversed. Note that the minimum temperature for alkylation in Frey, illustratively for benzene, is 70°F (approximately 21°C). Noting especially that Frey does not even disclose a hydrocarbon alkylation process for the compound of general formula (1) in claim 1, with each of R<sup>1</sup> and R<sup>2</sup> being a methyl group, it is respectfully submitted that the Examiner errs in her conclusion that one of ordinary skill in the art would have modified the temperature of alkylation to provide a temperature as in the present claims, for alkylation of the compound of general formula (1).

Assuming, arguendo, that the teachings of Olah and Frey were properly combinable; it is respectfully submitted that for simplification of processing, one of ordinary skill in the art would have conducted the alkylation in the copresence of the Broensted acid and the Lewis acid. In this regard, it is noted that Frey does not specifically disclose that the alkylation is to be conducted in the absence of a Lewis acid. To the contrary, Applicants have found that, according to the present invention, unexpectedly better results are achieved by conducting the alkylation in the presence of Broensted acid and without the presence of a Lewis acid, with a Lewis

acid subsequently being added and the isomerization taking place in the copresence of the Broensted acid and the Lewis acid. It is respectfully submitted that the unexpectedly better results achieved according to the present invention, as shown by the Examples and Comparative Examples in Applicants' disclosure, provide a basis for patentability of the presently claimed subject matter.

In view of the foregoing comments and amendments, reconsideration and allowance of all claims presently pending in the above-identified application are respectfully requested.

To the extent necessary, Applicants hereby petition for an extension of time under 37 CFR 1.136. Kindly charge any shortage of fees due in connection with the filing of this paper, including any extension of time fees, to the Deposit Account of Antonelli, Terry, Stout & Kraus, LLP, Account No. 01-2135 (case 396.44981X00), and please credit any overpayments to such Deposit Account.

Respectfully submitted,

**ANTONELLI, TERRY, STOUT & KRAUS, LLP**

By /William I. Solomon/  
William I. Solomon  
Registration No. 28,565

WIS/ksh  
1300 N. 17<sup>th</sup> Street, Suite 1800  
Arlington, Virginia 22209  
Tel: 703-312-6600  
Fax: 703-312-6666